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Fossil and modern sources of aerosol carbon in the Netherlands – A year-long radiocarbon study

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Measurement of the radioactive carbon isotope 14C in aerosols can provide a direct estimate of the contribution of fossil fuel sources to aerosol carbon. In aerosol science, measurements of 14C/12C ratios are usually reported as fraction modern (fm). The radiocarbon signature gives a clear distinction between 'modern' carbon sources (fm around 1.1-1.2 for biomass burning and around 1.05 for biogenic secondary organic aerosol) and 'fossil' carbon sources (fm =0 for primary and secondary formation from fossil fuel combustion). Due to the high cost of 14C analyses very few long-term studies have been conducted to date. The data that will be presented offer a unique insight into the seasonal variation of source contributions to the carbonaceous aerosol in a highly industrialized region.

High volume filter samples have been collected roughly twice per month from February 2011 – July 2012 at Cabauw, a rural location in the Netherlands surrounded by major urban centers and highways. This site provides a regional background aerosol contamination in the Netherlands. We report measurements of fm for total carbon (TC), organic carbon (OC), water insoluble OC (WIOC) and thermally refractory carbon (RC) as a proxy for elemental carbon.

The fraction modern of OC lies between 0.65 - 1 and shows only a moderate seasonal variation with highest values in the spring and lowest values in the summer. Elemental carbon is generally dominated by fossil fuel emissions, but shows a distinct seasonal variation with higher contribution of modern sources from November – Mai. This contribution is attributed to wood combustion. It is low when air masses arrive from the ocean and high for air masses with European continental origin, pointing to a main source outside the Netherlands. Water soluble organic carbon is dominated by modern sources throughout the year. For TC concentrations between 1.2 and 8 μg/m3, fm(TC) increases with TC concentration. A Keeling plot implies that synoptic scale variation in fm(TC) are mainly due to a modern source, imposed on a regional background with a relatively high contribution from fossil sources. TC concentrations > 8 μg/m3 are associated with pollution events transported from Germany/Eastern Europe and can have much high fossil contributions. However, fm in all carbon fractions is usually reduced in day time compared to night time, most likely due to traffic variations, which modify the carbonaceous aerosol on shorter time scales.

Using a simple model the contributions of fossil emissions, biomass burning, and biogenic emissions will be estimated for all carbon fractions. The seasonal variation in the source contributions will be presented and discussed.